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ELECTRICAL AND MAGNETIC PROPERTIES

# Effect of Chemically Active Medium on Frequency Dependence of Magnetic Losses in Soft Magnetic Fe-Based Amorphous Alloys

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**Abstract**—The effects of the electrolytic hydrogenation and oxidation and of the interaction of the surface ribbon with water and vapor on the frequency dependence of magnetic losses per magnetization-reversal cycle are studied based on the example of soft magnetic $Fe_{81}B_{13}Si_4C_2$ amorphous alloy, which exhibits a positive saturation magnetostriction. It was shown that, after the hydrogenation and oxidation of soft magnetic amorphous alloys, their frequency dependences of magnetic losses per magnetization-reversal cycle, which are reduced to unit induction, exhibit groups of hydrogen- and oxygen-related peaks in the frequency ranges of 35–55 and 55–80 Hz, which can be explained by the formation of O–*A* and H–*A* atomic pairs (where *A* are atoms of alloy components) and their reorientation in a magnetic field in the course of magnetization reversal at certain frequencies. The formation of analogous groups of peaks for samples of soft magnetic Fe-based amorphous alloys was observed after the interaction of the ribbon surface with water and vapor and after heat treatment in air. This fact confirms the possibility of the hydrogenation and oxidation of the alloys during the aforementioned processes.

*Keywords*: amorphous alloys, magnetic losses, heat treatment, electrolytic hydrogenation and oxidation

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Studies performed previously showed that the electrolytic surface saturation of soft magnetic amorphous alloys with hydrogen and oxygen affects the magnetization distribution in a ribbon via induction of mainly planar pseudo-uniaxial tensile stresses [1, 2]. This tension can be related to the anisotropic introduction of hydrogen and oxygen into the surface layer of ribbon due to the anisotropic magnetization distribution in the initial ribbon. Specific magnetic losses are one of important service characteristics of electrotechnical materials, since the losses determine the degree of efficiency of an article. Ribbons of soft magnetic Fe-based amorphous alloys are used over wide frequency and induction ranges. Therefore, the effect of various factors, in particular, electrolytic hydrogenation and oxidation and chemically active media at the ribbon surface on the frequency dependence of magnetic losses is of importance, and the problem calls for the investigation in detail. Media, which do not affect markedly the magnetization distribution and magnetic properties of soft magnetic crystalline alloys, can be chemically reactive for amorphous materials under the same conditions. The surface introduction of different atoms into an amorphous ribbon during a reaction with a chemically active medium leads to an increase in the concentration of diffusion-movable atoms and their pairs; this manifests itself in the character of the frequency dependence of magnetic losses [3–8]. In the present study, the effect of the electrolytic hydrogenation and oxidation, as well as the interaction of the ribbon surface with water and vapor on the frequency dependence of magnetic losses per magnetization-reversal cycle, was studied based on the example of soft magnetic $Fe_{81}B_{13}Si_4C_2$ amorphous alloy, which exhibits a positive saturation magnetostriction.

The studies were performed using samples in the form of strips 120 × 10 × 0.025 mm in dimensions, which were cut from an industrial ribbon produced at the JSC Asha Metallurgical Works. Magnetic losses were measured using the absolute wattmeter method in the regime of sinusoidal induction that was maintained using a feedback amplifier. The error of measurement of magnetic losses is ~4%.

Figure 1 demonstrates the effect of electrolytic hydrogenation and oxidation of the ribbon surface on the behavior of frequency dependence of magnetic losses per magnetization-reversal cycle for samples of the Fe–B–Si–C amorphous alloy subjected to heat treatment in air (at 380°C for 2 min), which leads to the formation of surface amorphous–crystalline layer of an optimum thickness. Saturation with hydrogen (oxygen) was performed using an electrolytic procedure, which consists of dipping the samples in an acid electrolyte and applying a current of a certain density. In this case, a sample saturated with hydrogen is the cathode, and a platinum wire is the anode; in the case of saturation with oxygen, the sample is the anode, and the platinum wire is the cathode. The comparison of data given in Figs. 1a and 1b shows that, immediately after the hydrogenation of the samples, the

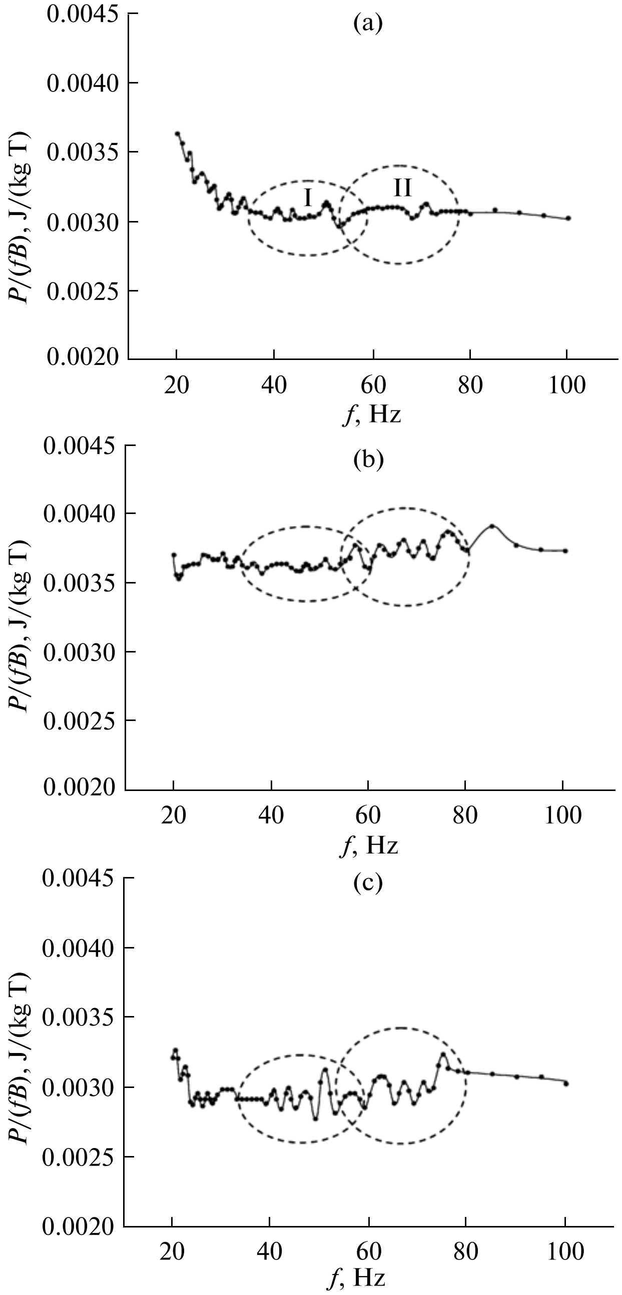


Fig. 1. Frequency dependence of magnetic losses per cycle of magnetization reversal ($B_m$ = 0.6 T) for samples of $Fe_{81}B_{13}Si_4C_2$ amorphous alloy preliminarily heat-treated in air at 380°C for 2 min: (a) initial state and after electrolytic saturation with (b) hydrogen and (c) oxygen.

frequency dependences exhibit a group of peaks in a frequency range of 55–80 Hz (region I).

The formation of these peaks is adequately explained by the theory of the directed ordering of atomic H–$A$ pairs that consist of different atoms, as well as by the reorientation of axes of atomic pairs when applying the magnetic field during magnetiza-

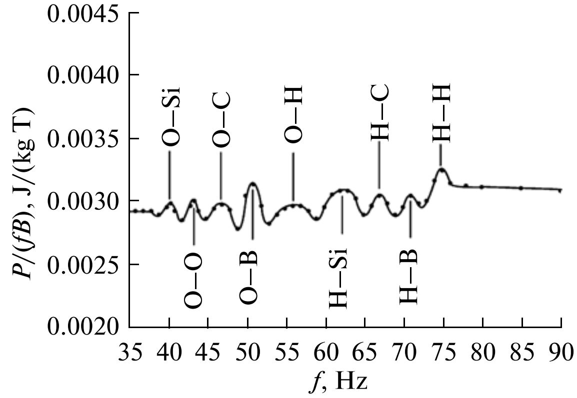


Fig. 2. Frequency dependence of magnetic losses per cycle of magnetization reversal for the $Fe_{81}B_{13}Si_4C_2$ amorphous alloy sample after electrolytic saturation with hydrogen (portion of Fig. 1; frequency range of 35–90 Hz.

tion and magnetization reversal [3–5]. The relaxation of axes of atomic pairs increases the viscosity field that decelerates the domain wall motion. This results in the intensification of their jump-like motion that favors the increase in magnetic losses [9]. The analogous results were obtained for the saturation of samples under study with oxygen (see Fig. 1c). Directly after oxidation, the frequency dependence of magnetic losses per magnetization-reversal cycle exhibits a group of peaks observed in a frequency range of 35–55 Hz (region I), which is also related to the appearance of pairs of different O–$A$ atoms, where $A$ = Si, O, C, B, and H. The fact that the series of peaks that correspond to the oxidation of the alloy is observed in the low the frequency range has drawn attention. This is related to the fact that the mass of oxygen atoms is higher than that of hydrogen atoms; therefore, the reorientation of the axes of O–$A$ atomic pairs takes a longer time (lower frequency) than the reorientation of the axes of H–$A$ atomic pairs. Note that the frequency dependence of magnetic losses per magnetization-reversal cycle exhibits a series of peaks related to hydrogen (II), along with the peaks related to oxygen (I). The existence of series II is due to the presence of water in the electrolyte, which, for the reasons described above, results in the hydrogenation of ribbon, along with its electrolytic oxidation. It is also remarkable that series I gradually approaches series II; therefore, a peak that covers both series must correspond to the reorientation of the axis of a pair of atoms equally related to both series, i.e., the O–H pair.

The results of studies have allowed us to tentatively conclude that there is a correspondence between an absorption peak and a certain pair of different elements, as well as to arrange them in order of increasing frequency and decreasing atomic mass of elements within a group as follows: (O–Si; O–O; O–C; O–B) O–H; (H–Si; H–C; H–B; H–H) (see Fig. 2, which is a fragment of Fig. 1c).

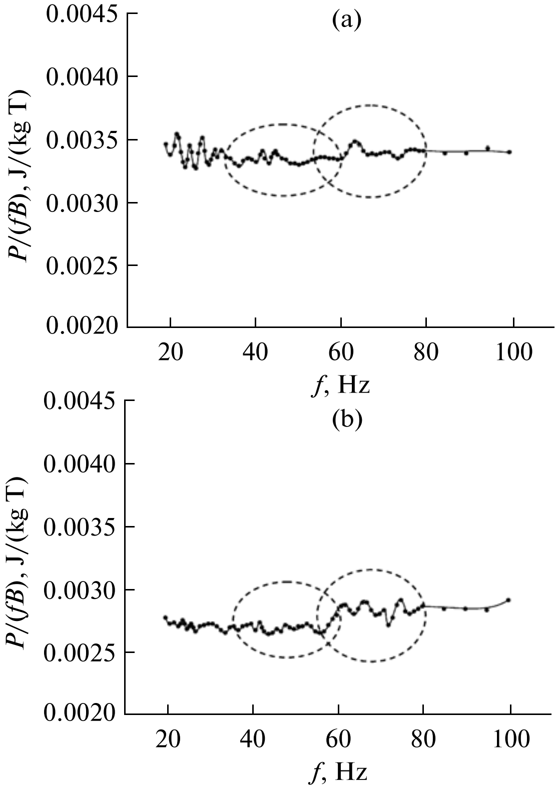


**Fig. 3.** Frequency dependence of magnetic losses per cycle of magnetization reversal ($B_m$ = 0.6 T) for samples of the $Fe_{81}B_{13}Si_4C_2$ amorphous alloy preliminarily heat-treated in air at 380°C for 2 min, which were measured 1 day after electrolytic saturation with (a) hydrogen and (b) oxygen.

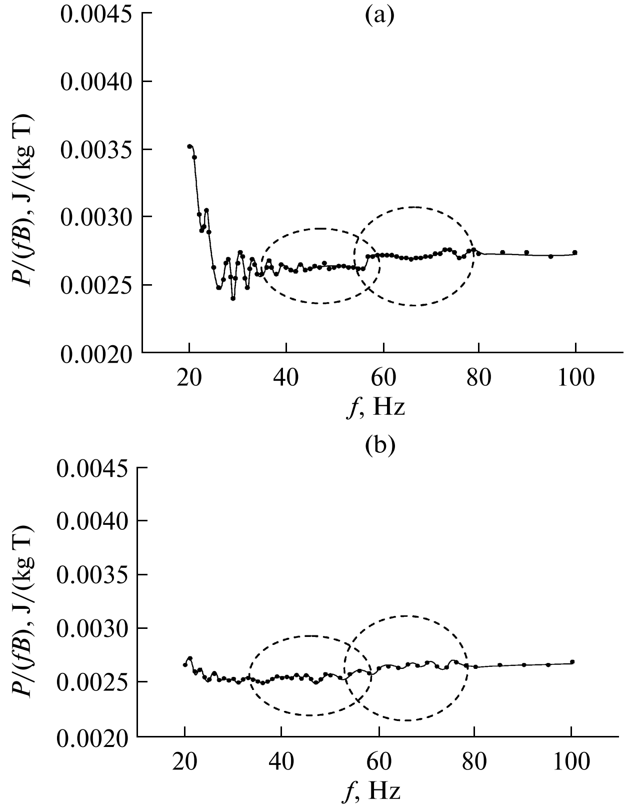


**Fig. 4.** Frequency dependence of magnetic losses per cycle of magnetization reversal ($B_m$ = 0.6 T) for samples of the $Fe_{81}B_{13}Si_4C_2$ amorphous alloy preliminarily heat-treated in air at 380°C for 2 min, which were measured 100 days after electrolytic saturation with (a) hydrogen and (b) oxygen.

The behavior of the frequency dependences of reduced magnetic losses for samples subjected to oxygen and hydrogen saturation changes over time (Figs. 3, 4). Even after 1 day, the intensity of absorption peaks observed in frequency ranges I and II decreases substantially, and peaks that correspond to O–O and H–H pairs disappear completely. This is related to the diffusion processes which lead to the outcome of hydrogen and oxygen from the ribbon. These results agree with the literature data of [10–12], according to which molecular hydrogen and oxygen escape from the matrix into the atmosphere and pores present in the matrix. Since silicon is a strong deoxidizer, peaks related to the reorientation of axes of O–Si and H–Si pairs are the most stable. This result also agrees with the literature data of [13]. Subsequently, due to the outcome of oxygen and hydrogen with time, the absorption peaks observed in the studied frequency range disappear progressively. Figures 4a and 4b demonstrate that, 100 days after hydrogenation, the peaks disappear almost completely, and the behavior of frequency dependence of the magnetic losses in a frequency range of 50–80 Hz is similar to that of the initial dependence.

Figure 5 shows the frequency dependence of magnetic losses per magnetization-reversal cycle for the as-quenched soft magnetic $Fe_{81}B_{13}Si_4C_2$ amorphous alloy and the alloy held in water for 1 min [14]. It can be seen that, at frequencies above 60 Hz, the magnitudes of magnetic losses are almost unchanged after holding in water. More substantial changes are observed for the low-frequency range, including a substantial decrease in the magnetic losses at a frequency of 20 Hz and the appearance (resolution) of an absorption peak at a frequency of ~30 Hz. The decrease in the magnetic losses at 20 Hz indicates a decrease in the activity of diffusion processes related to the diffusion of individual atoms. This can result from the hydrogenation and oxidation of the ribbon surface due to its reaction with water and the formation of new atomic pairs. As a result, the concentration of individual diffusion-movable atoms decreases, and the decrease in the viscosity field favors a decrease in the magnetic losses at a frequency of relaxation of individual atoms. The appearance of a peak at 30 Hz can be related to

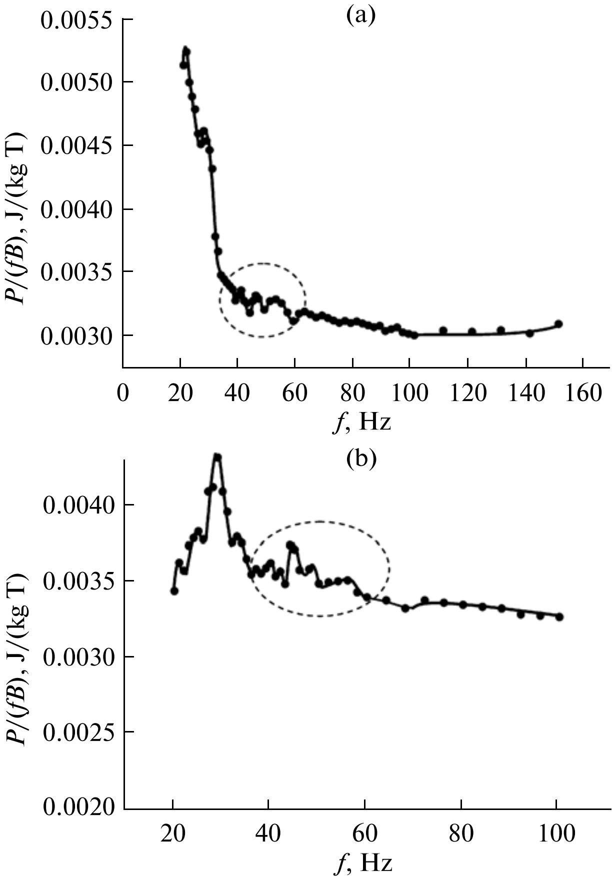


**Fig. 5.** Frequency dependence of magnetic losses per cycle of magnetization reversal at $B_m$ = 0.2 T for the $Fe_{81}B_{13}Si_4C_2$ amorphous alloy sample cut along the ribbon axis (a) in as-quenched state and (b) after 15 min holding in water.

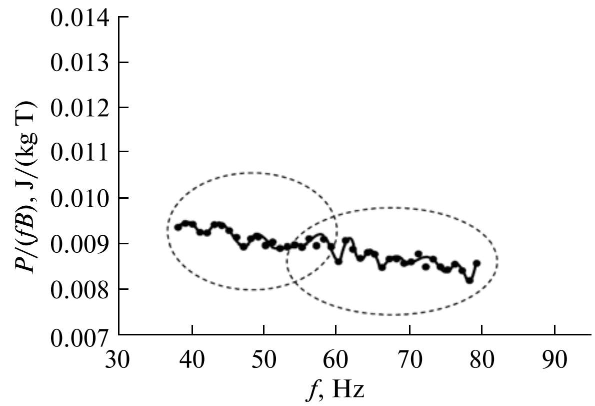


**Fig. 6.** Frequency dependence of magnetic losses per cycle of magnetization reversal at $B_m$ = 0.5 T for the $Fe_{81}B_{13}Si_4C_2$ amorphous alloy sample in the quenched state after 15 min treatment with vapor at room temperature.

the increasing role of the relaxation of axes of atomic pairs against the background of a decrease in the appearance of the relaxation of individual atoms. Figure 5 also demonstrates that weakly pronounced peaks of oxygen group are also observed for samples of quenched alloy. After dipping them in water, the intensity of oxygen-group peaks increases.

The effect of the vapor treatment of samples of the alloy ribbon at room temperature on the frequency dependence of magnetic losses (reduced to unit induction) per magnetization-reversal cycle is demonstrated in Fig. 6. It can be seen that, after vapor treatment, the frequency dependence of magnetic losses per magnetization-reversal cycle is characterized by groups of peaks, which are observed in the frequency ranges of 35–55 and 55–80 Hz. These peaks correspond to the oxygen and hydrogen groups and are analogous to those observed for samples of the studied ribbon alloy subjected to electrolytic oxidation and hydrogenation. Therefore, during the vapor treatment of the surface of an Fe-based amorphous alloy ribbon, its oxidation and hydrogenation take place in accordance with the oxidation reaction $2Fe + 3H_2O \rightarrow Fe_2O_3 + 3H_2\uparrow$.

Figure 7 shows the frequency dependences of magnetic losses per magnetization-reversal cycle, which were measured for the $Fe_{81}B_{13}Si_4C_2$ amorphous alloy ribbon directly after heat treatment in air at 380°C and 24 and 48 h after annealing. It can be seen that, after heat treatment in air, peaks of hydrogen and oxygen groups appear in a frequency range of 35–80 Hz. It can also be seen from Fig. 7a that the peak observed at a frequency of ~50 Hz exhibits the maximum intensity. In accordance with Fig. 2, this peak can be due to the formation of O–B atomic pairs; the degree of probability of the formation of these atomic pairs is fairly high, since the alloy under study is characterized by a high boron content. It also follows from Figs. 7b and 7c that, in this case, the intensities of peaks of the oxygen and hydrogen groups also change over time. Attention is drawn to the fact that the peak observed at ~50 Hz can only be measured directly after heat treatment. After 1 day, it becomes impossible to measure the magnetic losses in this frequency range due to the breakdown of wattmeter readings (the abrupt walkdown of the light cursor off the right-hand side of the scale), which is due to an abrupt increase in the magnetic losses within this frequency range. Changes in the intensities of peaks with time can be related to the diffusion of introduced atoms (hydrogen and oxygen) and atoms of other chemical elements, which are components of the amorphous alloy. Thus, these results confirm the assumption on the substantial effect of the oxidation and hydrogenation of the ribbon surface during heat treatment in air on the magnetization redistribution and magnetic properties of soft

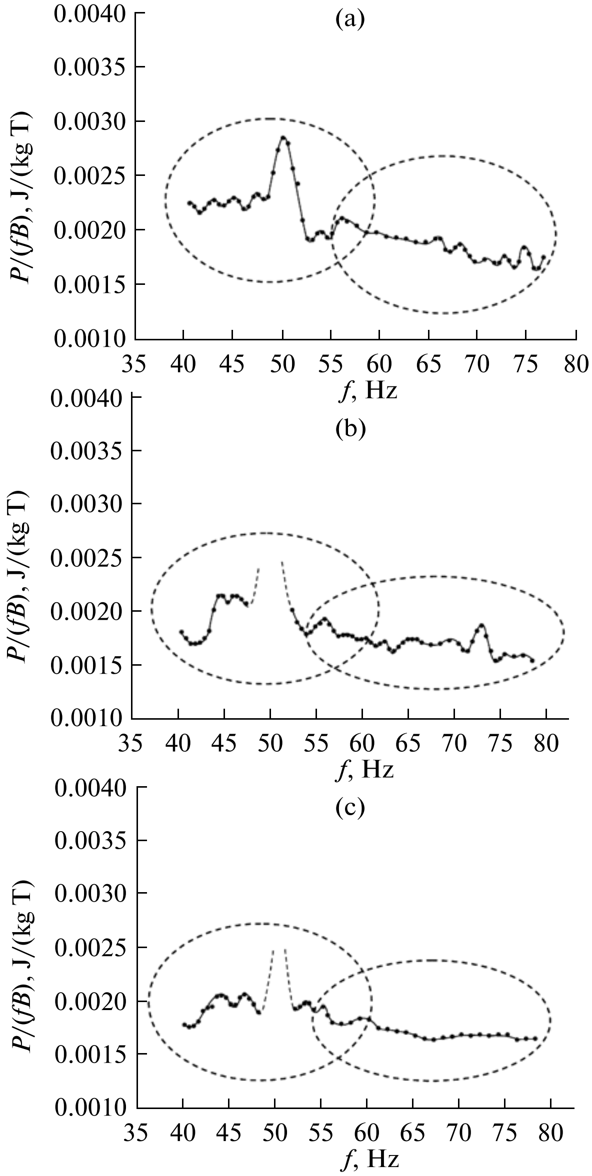


**Fig. 7.** Frequency dependences of magnetic losses per cycle of magnetization reversal of the $Fe_{81}B_{13}Si_4C_2$ amorphous alloy sample measured directly after (a) heat treatment in air at 380°C and (b) 1 and (c) 2 days after the treatment.

magnetic Fe-based amorphous alloys and the possibility of the occurrence of diffusion processes at room temperature.

## CONCLUSIONS

Results of performed studies show that the frequency dependence of magnetic losses (reduced to unit induction) per magnetization-reversal cycle for the soft magnetic alloys subjected to hydrogenation and oxidation demonstrates series of peaks of oxygen and hydrogen groups, which are observed in the frequency ranges of 35–55 and 55–80 Hz. These peaks can be explained by the formation of atomic pairs O–*A* and H–*A* (where *A* are atoms of chemical elements being the components of alloy) and their reorientation in the magnetic field applied in the course of magnetization reversal at certain frequencies. The formation of analogous groups of peaks was observed for ribbon of soft magnetic Fe-containing amorphous alloys that underwent surface interactions with water and vapor and were subjected to heat treatment in air. This confirms the possibility of hydrogenation and oxidation of the alloys during these processes.

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